Wednesday Morning, October 27, 1999

Surface Science Division Room 604 - Session SS3+EM-WeM

Surface Electronic Structure

Moderator: G.O. Sitz, The University of Texas, Austin

8:20am SS3+EM-WeM1 Spectroscopic Nonlinear Optical Responses of Clean, H-terminated and Ge-covered (2x1)Si(001), *L. Mantese*, *D. Lim*, *M.C. Downer*, University of Texas, Austin

Nonlinear optical spectroscopies are becoming important in elucidating the electronic structures of surfaces and interfaces. Yet, the accurate theoretical calculation of these spectra remains a difficult challenge. Recently, theoretical models have been developed to predict the nonlinear optical responses of clean and absorbate-covered (2x1)Si(001) surfaces.@footnote 1,2@ Here, we report the second harmonic (SH) responses of clean, H-terminated and Ge-covered (2x1)Si(001) to provide a direct test of predictive models. We have extended the range of previously reported results@footnote 3,4@ further into the IR where tunable IR femtosecond pulses in the range of ~1.15 to 2.5 microns were generated by a commercially available optical parametric amplifier (OPA) system. In this excitation region the SH response is resonantly enhanced by surface states including those from the dangling bonds and reconstruction-induced bonds. In addition, by directly using our oscillator output (700 to 800nm), we measure nonlinear optical responses in the E@sub 1@ region of Si. In this spectral range the SH response is highly sensitive and is altered in contrasting ways to ML coverages of H and Ge on (2x1)Si, originating from a surface electric field related to dimer charge transfer. The two measurement regions are discussed in comparison with theoretical predictions. @FootnoteText@ @footnote 1@B. Mendoza et al., PRL 81 (1998) 3781. @footnote 2@V.I. Gavrilenko et al., PRL submitted (1999). @footnote 3@U. Hofer, Appl. Phys. A 63 (1996) 533;J.I. Dadap et al., PRB 56 (1997) 13367. @footnote 4@P. Parkinson et al., Appl. Phys. B 68 (1999) 1.

8:40am **SS3+EM-WeM2 Si Surface Passivation: Si(111):GaSe versus Si(111):As, A. Bostwick,** S. Meng, B.R. Schroeder, University of Washington; E. Rotenberg, Lawrence Berkeley National Laboratory; F.S. Ohuchi, M.A. Olmstead, University of Washington

Heteroepitaxy on Si(111)7x7 requires removal of the deep reconstruction and accompanying dangling bonds. One way to passivate the Si(111) surface and remove the reconstruction is exposure to As, forming Si(111):As 1x1.@footnote 1@ The initial chemisorption of GeSe on Si(111)7x7 surfaces also results in a nearly ideally terminated 1x1 surface, quite similar to Si(111):As. Ga and Se occupy bulk Si sites, with Ga directly above Si (T1 site) and Se forming three back-bonds to Ga (H3 site). This (1x1) structure is the same as half a bulk GaSe layer, and initiates GaSe heteroepitaxy on Si(111). The very small (less than 0.1 eV) Si 2p core level shift shows the interface silicon to be in a bulk-like environment with minimal charge transfer, in contrast to As terminated silicon (0.75 eV shift). Electron counting arguments predict a lone-pair state on the Si(111):GaSe 1x1 surface, very similar to the As case. We observe such a state with angle-resolved ultraviolet photoemission spectroscopy. It has a similar E(k) dispersion to Si(111):As, though a somewhat larger bandwidth. We find a second surface state between this lone-pair state and the first bulk state, which we attribute to Ga-Se bonds. In addition, the zone-center bulk state, degenerate in bulk Si, is split by about 0.5 eV. This surprising result is not found for Si(111):As. We tentatively attribute the splitting to the Si-Ga interaction. @FootnoteText@ M. A. Olmstead, R. D. Bringans, R. I. G. Urhberg and R. Z. Bachrach, Phys. Rev. B 34, 6401 (1986).

9:00am SS3+EM-WeM3 Electron Excitations and Optical Properties of Semiconductor Surfaces, S.G. Louie, University of California, Berkeley and Lawrence Berkeley National Laboratory INVITED

The presence of electronic surface states and resonances often strongly modifies the structural, chemical, electronic, and optical properties of a surface. Over the past two decades, many sophisticated experimental methods (e.g., photoemission, scanning probes, and optical techniques) have been developed to investigate the spectroscopic properties of surfaces. In this talk, we discuss some of the theoretical developments in studying surface spectroscopic properties. Ab initio quasiparticle calculations can now be used to understand and predict surface-state excitation spectra such as those measured in photoemission and scanning tunneling spectroscopy experiments. A recent advance in treating excitonic effects further allows the first-principles study of surface optical processes. Results on several semiconductor surfaces will be presented. The possibility of using NMR chemical shifts as a probe of surface states will also be discussed.

9:40am SS3+EM-WeM5 Low Temperature Scanning Tunneling Microscopy and Spectroscopy of the @sr@3x@sr@3 R30° Phase of Sn/Si(111), *L. Ottaviano*, INFM, Italy; *A. Continenza, M. Crivellari, L. Lozzi,* University of L'Aquila and INFM, Italy; *S. Modesti,* University of Trieste and Laboratorio TASC-INFM, Italy; *S. Picozzi, G. Profeta, S. Santucci,* University of L'Aquila and INFM, Italy

In these years there has been a great deal of efforts in the understanding of the low temperature transition from a @sr@3x@sr@3 R30° to a 3x3 phase of Sn(Pb)/Ge(111).@footnote 1@,@footnote 2@ On the other hand, little attention has been devoted to the investigation of iso-electronic interfaces like Sn(Pb)/Si(111) at similarly low temperatures. Among other experimental techniques, STM can provide a deep understanding of the electronic structure of these surfaces by performing measurements at various tunnelling voltages. Theorist who proposed different model interpretations have recently called for STM experiments at low temperatures on such systems.@footnote 3@ We show the first STM atomically resolved images of the low temperature (100 K) 1/3 ML-Sn/Si(111) @sr@3x@sr@3 R30° phase. Correspondingly we have measured tunnelling I-V spectra. Data are compared with all electron ab-initio calculations performed on a slab of Si(111) covered with 1/3 ML-Sn in the @sr@x@sr@3 R30° reconstruction. In particular we compare the ab-initio results with surface and electronic properties as obtained by STM/STS and photo-emission results. @FootnoteText@ @footnote 1@ L.Petaccia, L.Grill, M. Zangrando, and S. Modesti, Phys. Rev. Lett. 82 (1999) 386. @footnote 2@ A. Mascaraque et al., Phys. Rev.Lett. 82 (1999) 2524. @footnote 3@ S. Scandolo, F. Ancilotto, G.L.Chiarotti, G. Santoro, S. Serra, and E.Tosatti, Surf. Sci. 402-404 (1998) 808.

10:00am SS3+EM-WeM6 Electronic Structure of the 3C-SiC(001)-c(4x2) Surface, L. Duda, L.S.O. Johansson, B. Reihl, Universität Dortmund, Germany; H.W. Yeom, University of Tokyo, Japan; S. Hara, S. Yoshida, Electrotechnical Laboratory Tsukuba, Japan

We provide the first investigations of the electronic structure of the Si-rich 3C-SiC(100)-c(4x2) surface using angle-resolved photoemission and synchrotron radiation. The surface is found to be semiconducting. The Fermi level position was determined to be at 1.9 eV above the valenceband maximum. Three surface states are identified within the bulk band gap. We measured the dispersion of these states along the high-symmetry directions of the surface Brillouin zone (SBZ). The upper band at 1 eV binding energy and the third band at 2.6 eV are non-dispersive. The second band is located at 1.5 eV at the center of the SBZ and shows a weak dispersion of 0.3 eV in the @Gamma@-X direction, whereas, it is dispersionless in the other directions. We compare these results to calculated band structures.@footnote 1@ In addition, the comparison with experimental observed band structures of the 2x1 surface@footnote 2@ gives important new insight into the reconstruction behaviour of the SiC surface, supporting the close similarity of both reconstructions. @FootnoteText@ @footnote 1@ W. Lu, P. Krüger, J. Pollmann, Phys. Rev. Lett. 81 (1998) 2292 @footnote 2@ L. Duda, L. S. O. Johansson, B. Reihl, H. W. Yeom, S. Hara, S. Yoshida, submitted to Surface Science (1999).

10:20am SS3+EM-WeM7 Photoemission Spectroscopy of Platinum Overlayers on Silicon Dioxide Films, J.W. Keister, J.E. Rowe, Army Research Office; J.J. Kolodziej, T.E. Madey, Rutgers University

Soft x-ray photoelectron spectroscopy has been used to study ultrathin Pt films on silicon dioxide as model supported-catalyst materials. Using monochromatic synchrotron radiation (NSLS U4A - Brookhaven), platinum and silicon core-level photoelectron peaks were measured as a function of platinum coverage in the range, 0 - 10 monolayers (ML). The bulk Si and SiO@sub 2@ film Si(2p) peaks both show a binding energy drop within the first ~ 0.5 ML of dosing. However, the effect is stronger for the SiO@sub 2@ film Si(2p) peak, indicating an increased screening of the 2p electrons by the metal overlayer. We also observe a monotonic increase of the Pt film workfunction with coverage from 4.52 eV initially to 5.59 eV at ~ 10 ML. The Pt(4f) and valence band spectra display changes with Pt dose which are also consistent with increased screening, from non-metallic below ~ 0.5 ML to metallic as coverage increases. The Pt core level decreases its binding energy from ~ 72.2 eV to ~ 70.9 eV between 0 and 10 ML coverage. The coverage dependence of the Pt lineshape and intensity, and the large saturation coverage for the workfunction change indicate cluster growth (Volmer-Weber growth). The increasingly metallic nature of the Pt overlayer with dose is quantified by numerical lineshape fitting with

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Gaussian-broadened Doniach-Sunjic functions, as appropriate for varying amounts of metallic screening. Further quantitative results and interpretation in terms of nucleation and growth of Pt clusters will be discussed.

10:40am SS3+EM-WeM8 The Behavior of Metallic Quantum Well State Dispersions in the Cu/fccCo/Cu(100) System as a Function of Film Thickness, W.K. Siu, R.A. Bartysnki, Rutgers University

The magnetic coupling of ferromagnetic (FM) layers separated by a nonmagnetic (NM) layer has been associated with the formation and behavior of metallic quantum well (MQW) states in the spacer layer. Inverse photoemission has been used to investigate the MQW states in the prototypical Cu/fccCo/Cu(100) system for very thin Cu layers. These studies have shown that their dispersion with parallel momentum can be strongly affected by the electronic structure of the underlying FM material. Specifically, rapidly dispersing MQW states in the NM layer will acquire an uncharacteristically large effective mass when they encounter a projected band gap in the FM material. The experimental result shows the the behavior over severval Cu monolayers. In the Cu/fccCo/Cu(100) case, this occurs near the neck of the Cu Fermi surface and affect the states responsible for the short period coupling in this system. We have investigated how the dispersion of these levels develops as a function of Cu layer thickness and, in particular, how it evolves towards the behavior found for the single crystal Cu(100) surface. These results are explained in terms of a phase accumulation model for the electronic structure of the MQW system. Funded by the National Science Foundation under grant No. NSF-DMR98-01681. and the Petroleum Research Fund under grant No. ACS-PRF-33750-AC6,5.

11:00am SS3+EM-WeM9 Atomic and Electronic Structure of Ag/Ni(110) and Ni/Ag(100), P.T. Sprunger, D.A. Hite, Louisiana State University

The atomic and electronic structure of Ag on Ni(110) and Ni on Ag(100) has been studied by synchrotron-based angle-resolved photoemission spectroscopy (ARUPS), low energy electron diffraction and scanning tunneling microscopy. These systems are of interest due their high bulk lattice mismatch and immiscibility. STM results of the Ag/Ni(110) system have revealed a novel pseudomorphic, surface-confined alloy phase at a coverage of ~0.5 ML. ARUPS results of this phase show a bulk-like, 3-D Ag electronic structure due to strong hybridization with the Ni bands. However, at a coverage of 1 ML, the Ag "de-alloys" and structurally forms a strained ~ Ag(111) overlayer. The electronic structure of this phase is radically different, namely, the laterally localized Ag d-bands show only kparallel dispersion, that is, 2-D structure. In contrast, ARUPS results of Ni monolayer deposition on Ag(100) reveals no alloy formation below 150 K. However, at deposition temperatures above 400 K, Ni segregates subsurface, resulting in a Ag surface termination. The underlying Ni adopts Ag lattice parameters. ARUPS data from this latter phase reveals 3-D, bulklike electronic structure due to strong mixing of states. The results will be compared and contrasted with theoretical band-structure calculations. This research is funded by DOE through contract DE-FG-02-98ER45712.

11:20am SS3+EM-WeM10 Spin-Orbit-Split Surface States in Clean and Alkali-Metal-Covered W(110)@footnote 1@, E. Rotenberg, Lawrence Berkeley National Laboratory; J.W. Chung, Pohang Institute of Science and Technology, Korea; S.D. Kevan, University of Oregon

The broken symmetry at surfaces can give rise to a non-zero spin-orbit splitting of valence bands, as has recently been observed for the s-p derived surface state on Au(111).@footnote 2@ We now observe such a splitting of the d-derived surface state on W(110) and to a lesser extent, on Mo(110), and follow the evolution of the splitting as monovalent atoms are adsorbed. We find a dramatic increase in the splitting as a function of deposition for H and all alkali metals for coverages up to 1 ML. For Li, which shows the strongest change in splitting, we find a decrease in the splitting at the highest coverages near 1 ML. Our findings show that the spin-orbit splitting is a sensitive probe of the local surface conditions, to the extent that they are sampled by the surface state's wavefunction. The observed evolution is also directly relevant to recent observations of altered orbital magnetic structure vs. adsorbate coverage in magnetic materials. We propose a spin ordering for the associated Fermi contours which has important implications for spin excitations at surfaces. @FootnoteText@ @footnote 1@E. Rotenberg, J. W. Chung, and S. D. Kevan, Phys. Rev. Lett. 82 4066 (1999). @footnote 2@S. LaShell, B. A. McDougall, and E. Jensen, Phys. Rev. Lett., 3419 (1996).

11:40am SS3+EM-WeM11 Resonant Auger Studies of Metallic Systems, I. Coulthard, J.W. Freeland, S.P. Frigo, J.F. Moore, W.S. Calaway, M.J. Pellin, M. Mendelsohn, Argonne National Laboratory; T.K. Sham, S.J. Naftel, University of Western Ontario, Canada; A. Stampfl, Australian Nuclear Science and Technology Organization

Auger electron emission at photon energies near the associated absorption edge which generates the initial core hole is a complex process. Excitation and decay become linked such that the process must be described as a single-step process rather than the classical two step process for Auger emission. This results in the production of discrete lines which are resonantly enhanced at the absorption edge, and whose intensity vanishes far below or above the edge. Another result of utilizing resonant photon energies is that the Auger line width may no longer be limited by the intrinsic lifetime resulting in a narrowing of the Auger spectrum.@footnote 1@ Results will be presented for a variety of systems including. Cu. Co. Al. CuNi alloys, and CuAl alloys. Specific attention will be paid to:probing unoccupied densities of states, very high resolution x-ray absorption spectroscopy utilizing auger yields to produce sub-lifetime narrowed x-ray absorption spectra, and the use of the technique for non-homogeneous sample systems. Work at Argonne is supported by the U.S. Department of Energy, Basic Energy Sciences, under Contract No. W-31-109-Eng-38. @FootnoteText@@footnote 1@T.M. Grehk, W. Drube, R. Treush, and G. Materlik. Phys. Rev. B 57, 6422 (1998).

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